

REMARKS

Applicant has added new claims 19 and 20. Support for these two new claims can be found in claim 1. No new matter has been introduced by the above amendment. Claims 1-20 are currently pending. Reconsideration of the application, as amended, is respectfully requested in view of the remarks below.

I

Claims 1-12 and 16-18 are rejected under 35 U.S.C. 103(a) as being obvious over Kato et al., EP 0365308 A2 ("Kato") in view of Tamura et al., U.S. Patent 5,041,272 ("Tamura") and Hayasaka et al., U.S. Patent 5,312,608 ("Hayasaka"). See the Office Action, page 2, lines 14-16.

Independent claims 1 and 16 will be discussed first. Claim 1 is drawn to an exhaust gas purifying catalyst, which includes (1) a composite oxide and (2) a zeolite. The composite oxide contains zirconium, manganese, and, optionally, cobalt. The zeolite is a proton zeolite, or a zeolite modified with cerium, lanthanum, phosphorus, boron, gallium, magnesium, or a mixture of these elements. Claim 16 is drawn to a method for purifying various nitrogen oxides in an exhaust gas using a catalyst of claim 1, in which the composite oxide and the zeolite is physically mixed.

The Examiner correctly points out that "Kato et al. discloses an exhaust gas purifying catalyst wherein the catalyst comprises Co supported by Zr oxide mixed with a zeolite (see abstract)." See the Office Action, page 2, lines 21-22. The Examiner then proceeds to conclude that "it would have been obvious to one of ordinary skill in the art to modify the teachings to Kato et al. based on the teachings of Tamura et al. and Hayasaka et al., ... because Tamura et al. teaches a method ... by contracting the catalyst with a hydrogenated zeolite, which is impregnated with one or more kinds of metals and Hayasaka et al. ... teach[es] that there is no particular restriction on the kind of zeolite" See the Office Action, page 3, lines 12-18. Applicant disagrees.

Kato discloses a catalyst containing (1) Cu and/or Co supported by Zr oxide and (2) a Cu-substituted zeolite. See, e.g., the Abstract and page 2, lines 38-39. However, it does not disclose or suggest either (1) a composite oxide containing zirconium and manganese or (2) a proton zeolite or a zeolite modified with cerium, lanthanum, phosphorus, boron, gallium, magnesium, or a mixture of these elements, both required by claims 1 and 16.

Tamura discloses hydrogenated zeolite catalysts or hydrogenated zeolite catalysts impregnated with one or more metals. See, e.g., the Abstract and column 1, lines 63-64. It is silent on use of a composite oxide, let alone use of a composite oxide containing zirconium and manganese required by claims 1 and 16. Thus, Tamura does not cure the deficiency in Kato, which discloses Cu and/or Co supported by Zr oxide, but not a composite oxide containing zirconium and manganese as required by claims 1 and 16.

Hayasaka discloses a catalyst having (1) "gallium as a main catalyst," (2) at least one [metal] selected from iron, nickel, cobalt, zirconium, manganese, chromium, molybdenum, copper, cerium, titanium, niobium ... as a promoter," and (3) "a zeolite." See, e.g., the Abstract and column 3, lines 7-11. The gallium source can be any compounds that are convertible into oxides, e.g., gallium oxide. See column 3, lines 26-29. While Hayasaka explicitly states that "the catalyst containing gallium as a main catalyst," claims 1 and 16 cover a catalyst having a composite oxide that is required to contain zirconium and manganese, rather than gallium. Thus, Hayasaka also does not cure the deficiency in Kato.

Thus, claims 1 and 16 are not rendered obvious by Kato in view of Tamura and Hayasaka. In other words, the Examiner has failed to establish a *prima facie* case of obviousness.

Even if a *prima facie* case of obviousness has been made (which Applicant does not concede), it can be successfully rebutted by a showing of an unexpected advantage of the exhaust gas purifying catalyst recited in claims 1 and 16. See Mr. Tatsuya Yoshikawa's Declaration ("Declaration"), a copy of which is attached hereto as "Exhibit A."

The Declaration describes a number of experiments. In Experiment 1, a claimed catalyst was prepared and tested. In Comparative Experiments 2, 4, and 5, three prior art catalysts were prepared and tested¹. Specifically, Experiment 1 describes a catalyst recited in claims 1 and 16, which contains oxides of manganese and zirconium, and H-ZSM5 (a proton zeolite). Comparative Experiment 2 describes a catalyst disclosed in Kato, i.e., a catalyst containing oxides of copper and zirconium, and Cu-ZSM5 (a zeolite modified with copper). Comparative

¹ Experiment 2 described in the Declaration will be discussed below. The results described in Comparative Example 1 and 6-8 are similar to those of Comparative Experiments 2 and 5, respectively. Thus, these results are not discussed herein.

Experiment 4 describes a catalyst disclosed in Hayasaka, i.e., a catalyst containing gallium and ZSM5. Comparative Experiment 5 describes a catalyst disclosed in Tamura, i.e., a catalyst containing H-ZSM5 (a proton zeolite). The catalyst described in Experiment 1 shows a much higher NO_x conversion rate (55%) than those of the catalysts described in Comparative Experiment 2 (36%), Comparative Experiment 4 (12%), and Comparative Experiment 5 (11%). See Table B of the Declaration. Given this unexpected advantage, claims 1 and 16 are clearly not rendered obvious by Kato in view of Tamura and Hayasaka.

As claims 2-12 and 17-18 depend from claims 1 and 16, they are also not rendered obvious by Kato in view of Tamura and Hayasaka.

II

Claims 13-15 are rejected under 35 U.S.C. 103(a) as being obvious over Kato in view of Tamura and Hayasaka, and further in view of Montreuil et al., U.S. Patent 5,238,672 ("Montreuil"). See the Office Action, page 4, lines 3-6.

Each of claims 13-15 is drawn to a catalyst of claim 1, which has two components. The first component (i.e., a composite oxide) further contains at least one element selected from the group consisting of bismuth, iron, cerium, praseodymium, gadolinium, lanthanum, barium, strontium, calcium, cesium, yttrium, and mixtures thereof. The second component (i.e., a zeolite) is a proton zeolite, or a zeolite modified with at least one element selected from the group consisting of cerium, lanthanum, phosphorus, boron, gallium, magnesium, and mixtures thereof.

As discussed above, neither Kato nor Tamura discloses or suggests a catalyst having a composite oxide containing zirconium and manganese, which is required by each of claims 13-15. Further, Hayasaka teaches that gallium is an essential component in the oxide phase of a catalyst. Thus, it does not suggest a catalyst of claims 13-15, in which zirconium and manganese, rather than gallium, are essential components in the oxide phase.

Turning to Montreuil, the Examiner points out that this reference discloses "a dual-phase catalyst having a transition metal-containing zeolite phase and a transition metal-containing oxide phase. The oxide phase is comprised of zirconium oxide and transition metals, such as manganese, copper, cobalt, iron, calcium, and mixtures thereof." See the Office Action, page 4, lines 13-16. The Examiner then proceeds to conclude that "it would have been obvious to one of

ordinary skill in the art to modify the teachings of Kato et al., Tamura et al., and Hayasaka et al., based on the teachings of Montreuil.” See the Office Action, page 4, lines 19-21. Applicant disagrees.

Montreuil discloses a catalyst in which the transition metals in the zeolite and oxide phases can be copper, cobalt, nickel, chromium, iron, manganese, silver, zinc, and calcium. See column 2, lines 19-23. Montreuil explicitly states that “[t]he transition metal[s] present in the two materials [i.e., the oxide and zeolite phases] of this catalyst preferably are the same ... [p]referably, this transition metal is copper.” See column 2, lines 23-27; emphases added. In view of this statement, Montreuil clearly does not suggest a catalyst covered by each of claims 13-15, in which (1) the composite oxide contains metals that are not copper and are not the same as those, if any, contained in the zeolite; and (2) the zeolite contains no transition metal or contains a metal that is not copper or any of the other metals disclosed in Montreuil. Indeed, the catalyst of claims 13-15 does not contain copper at all.

Thus, claims 13-15 are not rendered obvious by Kato in view of Tamura and Hayasaka, and further in view of Montreuil. In other words, the Examiner has failed to establish a *prima facie* case of obviousness.

Even if a *prima facie* case of obviousness has been made (which Applicant does not concede), it can be successfully rebutted by a showing of an unexpected advantage of an exhaust gas purifying catalyst covered by claims 13-15. See the Declaration. Specifically, in Experiment 2 described in the Declaration, a catalyst covered by each of claims 13-15, i.e., containing oxides of manganese, cerium, and zirconium, and H-ZSM5 (a proton zeolite), was prepared and tested. In Comparative Experiment 2 described in the Declaration, a catalyst disclosed in Montreuil, i.e., containing oxides of copper and zirconium, and Cu-ZSM5 (a zeolite modified with copper), was prepared and tested. The catalyst described in Experiment 2 shows a much higher NO_x conversion rate (75%) than that of the catalyst described in Comparative Experiment 2 (36%). See Table B of the Declaration. Given this unexpected advantage, claims 13-15 are clearly not rendered obvious by Kato in view of Tamura and Hayasaka, and further in view of Montreuil.

Applicant : Tatsuya Yoshikawa
Serial No. : 09/888,009
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Attorney's Docket No.: 13298-004001 / F 2001-43-US

CONCLUSION

Applicant submits that the grounds for rejection asserted by the Examiner have been overcome, and that claims 1-20, as pending, define subject matter that is non-obvious. On this basis, it is submitted that all claims are now in condition for allowance, an action of which is requested.

Please apply any other charges to deposit account 06-1050, referencing Attorney's Docket No. 13298-004001.

Respectfully submitted,

Date: 1-12-04

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICERECEIVED
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TC 1700

In re application of Tatsuya Yoshikawa

Serial No.: 09/888,009

5 Filed: June 22, 2001

Title: EXHAUST GAS PURIFYING CATALYST

Art Unit: 1754

Examiner: JONAS N STRICKLAND

10 COMMISSIONER FOR PATENTS
WASHINGTON, D. C. 20231DECLARATION UNDER 37 C. F. R. 1.132

Sir:

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I. I, Tatsuya YOSHIKAWA, of b-612, 1736-3, Imajuku, Himeji, Hyogo 670-0052, Japan, hereby declare that I am an inventor named in the above-mentioned application, that I graduated from Kyoto University, Department of Engineering and
20 Petroleum Chemistry in March 1988 with a B.S. degree. I received my master's degree, molecular engineering, from the same University in March 1990, that, since April 1990, I have been employed by Nippon Shokubai Co., Ltd., and engaged in exhaust gas purifying catalyst research for last 7 years.
25 In the period, I have been to Reading University, UK, for 2 years as guest researcher to study surface science from 1994 to 1996. Additionally, in accordance with our section consolidation to ICT Co., Ltd, I have been in temporal transfer to ICT Co., Since 1998.

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II. I understand that claims 1-12 and 16-18 of the above applicati n have been reject d under 35 U. S. C. 103 (a) as

being unpatentable over Kato et al. (EP 0365308 A2) in view of Tamura et al. (US Patent 5,041,272) and Hayasaka et al. (US 5,312,608), that claims 13-15 have been rejected under 35 U.S.C. 103(a) as being unpatentable over Kato et al. (EP 0365308 A2) in view of Tamura et al. (US Patent 5,041,272) and Hayasaka et al. (US 5,312,608) as applied to claims 1-12 and 16-18 above, and further in view of Montreuil et al. (US Patent 5,238,672) and I have reviewed these references. It is of my opinion that the claimed catalysts in the present application are nonobvious over the cited references because the claimed catalyst achieved superior results as shown by the following experimental results.

EXPERIMENT 1 (corresponding to Example 1 of the present specification)

One thousand (1000) g of zirconium oxide was impregnated with an aqueous solution containing a 100 g equivalent of manganese nitrate as manganese oxide (MnO_2), dried overnight at 120°C, and calcined at 550°C for one hour. The resultant manganese-deposited zirconium oxide and H-2SM5 (Si/Al = 27, available from Zeolyst International) were separately ball milled in an aqueous solution, and mixed at a weight ratio (solid ratio) of 2: 8. The produced mixture was applied to a monolith made of cordierite (400 cells, 0.043 liter, available from NGK Insulators, LTD in Japan) so as to account for an amount of 180 g/L. The coated monolith thus obtained was dried at 120°C and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table 1.

EXPERIMENT 2 (corresponding to Example 2 of the present specification)

One thousand (1000) g of zirconium oxide was impregnated with an aqueous solution containing a 100 g equivalent of manganese nitrate as manganese oxide (MnO_2) and a 40 g equivalent of cerium nitrate as cerium oxide (CeO_2), dried overnight at 120°C , and calcined at 550°C for one hour. The resultant manganese-deposited zirconium oxide and H-2SM5 (Si/Al = 27, available from Zeolyst International) were separately ball milled in an aqueous solution, and mixed at a weight ratio (solid ratio) of 2: 8. The produced mixture was applied to a monolith made of cordierite (400 cells, 0.043 liter, available from NGK Insulators, LTD in Japan) so as to account for an amount of 180 g/L. The coated monolith thus obtained was dried at 120°C and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table 1.

In the reaction gas shown in Table 1, the produced catalyst was heated from 100°C to 550°C at an increasing rate of $20^\circ\text{C}/\text{min}$. and cooled from 550°C to 150°C at a decreasing rate of $20^\circ\text{C}/\text{min}$. to determine conversion efficiencies of NO_x .

TABLE 1. CONDITIONS OF REACTION GAS

Conditions for rating model gas	
NO: 400 ppm,	
Propylene: 1800 ppm C_1 (normalized to carbon concentration), CO: 400 ppm, O_2 : 8.5 vol %, Water: 10 vol %	
Total flow volume: 28 L/min., SV: About 40,000 h^{-1}	

NO was analyzed with a chemical emission spectral analyzer (CLD), and the hydrocarbon with a hydrogen flame ionizing analyzer (NDIR), respectively.

The result obtained is shown in Table B below.

COMPARATIVE EXPERIMENT 1

A catalyst was prepared according to the procedure of Experiment 1 while using Cu-Mordenite (Example 1 in Kato et al.) instead of H-ZSM5. The catalyst performance was measured under the conditions in Table 1 and the result obtained is shown in Table B below.

COMPARATIVE EXPERIMENT 2

A catalyst was prepared according to the procedure of Experiment 1 while using Cu-ZSM5 (Example 1 in Montreuil et al.) instead of H-ZSM5 and using CuOx/ZrO₂ (Example 1 in Montreuil et al. and also Kato et al.) instead of MnOx/ZrO₂. Cu content in the CuOx/ZrO₂ powder was 10wt.% as CuO. The catalyst performance was measured under the conditions in Table 1 and the result obtained is shown in Table B below.

COMPARATIVE EXPERIMENT 4

Ga-aluminosilicate and silica sol (binder) was ball milled 14 hours in an aqueous solution with the ratio of 85% and 15% as solid amount. The resultant slurry was coated on a monolith made of cordierite (400 cells, 0.043 liter) so as to account for an amount of 180g/L. The coated monolith thus obtained was dried at 120°C and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table 1.

The reaction gas condition is shown in Table 1. The produced catalyst was heated from 100°C to 550°C at an increasing rate of 20°C/min. and cooled from 550°C to 150°C at a decreasing rate of 20°C/min. to determine conversion efficiencies of NOx. The NOx conversion was measured in the

decreasing temperature run.

COMPARATIVE EXPERIMENT 5

H-ZSM5 (Si/Al=27 available from Zeolyst International)
5 and silica sol (binder) was ball milled 14 hours in an aqueous solution with the ratio of 85% and 15% as solid amount. The resultant slurry was coated on a monolith made of cordierite (400 cells, 0.043 liter) so as to account for an amount of 180 g/L. The coated monolith thus obtained was dried at 120°C
10 and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table 1.

The reaction gas condition is shown in Table 1. The
15 produced catalyst was heated from 100°C to 550°C at an increasing rate of 20°C/min. and cooled from 550°C to 150°C at a decreasing rate of 20°C/min. to determine conversion efficiencies of NOx. The NOx conversion was measured in the decreasing temperature run.

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COMPARATIVE EXPERIMENT 6

A catalyst was prepared according to the procedure of Comparative experiment 1 while using H-Mordenite (Si/Al=40) instead of H-ZSM5. The catalyst performance was measured
25 under the conditions in Table 1.

COMPARATIVE EXPERIMENT 7

A catalyst was prepared according to the procedure of Comparative experiment 1 while using H-beta zeolite
30 (Si/Al=50) instead of H-ZSM5. The catalyst performance was measured under the conditions in Table 1.

COMPARATIVE EXPERIMENT 8

A catalyst was prepared according to the procedure of Comparative experiment 1 while using H-ZSM5 (Si/Al=80) instead of H-ZSM5 (Si/Al=27). The catalyst performance was measured under the conditions in Table 1.

TABLE B

	Catalyst		NOx conv. (%) (HC/NOx=4.5)
	1st Component	2nd Component	
Experiment 1	MnOx/ZrO ₂	H-ZSM5 (Si/Al=27)	55
Experiment 2	MnCeOx/ZrO ₂	H-ZSM5 (Si/Al=27)	75
Comparative Experiment 1	MnOx/ZrO ₂	Cu-Mordenite	28
Comparative Experiment 2	CuOx/ZrO ₂	Cu-ZSM5	36
Comparative Experiment 4	NONE	Ga-aluminosilicate (ZSM5 structure)	12
Comparative Experiment 5	NONE	H-ZSM5 (Si/Al=27)	11
Comparative Experiment 6	NONE	H-Mordenite (Si/Al=40)	7
Comparative Experiment 7	NONE	H-beta zeolite (Si/Al=50)	13
Comparative Experiment 8	NONE	H-ZSM5 (Si/Al=80)	13

CONCLUSION

10 It becomes clear from the above results, the present invention provided superior NOx conversion to the catalysts of Comparative Experiments 1, 2 and 4-8, which are derived

from Kato et al., Montreuil et al., (Comparative experiments 1 and 2), Hayasaka et al., (Comparative experiment 4), and Tamura et al., (Comparative experiments 5-8).

- 5 III. The undersigned declares that all statements made herein are true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fines or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that
10 such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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7th Jan. 2004

Date

Tatsuya Yoshikawa

Tatsuya YOSHIKAWA